Low-frequency dielectrophoretic response of a single particle in aqueous suspensions

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We use optical tweezers-based dielectrophoresis (DEP) force spectroscopy to investigate the roles of the electrical double layer in the AC dielectric response of an individual colloidal particle in an aqueous medium. Specifically, we measure the DEP crossover frequency as a function of particles size, medium viscosity, and temperature. Experimental results were compared to low frequency relaxation mechanisms predicted by Schwarz, demonstrating the dielectrophoretic responses in the frequency range between 10 kHz and 1 MHz were dominated by counterion diffusion within the electric double layer. © *2016 AIP Publishing LLC*.
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I. INTRODUCTION

Dielectrophoresis (DEP), an electrically driven motion of polarizable particles in a non-uniform electric field, is a phenomenon reported by Pohl almost four decades ago.[1–3] Different from the more well-known electrophoresis, DEP force depends on the relative polarizability of the particle in the suspending medium, and the magnitude of the force is proportional to ∇E^2. When the polarizability of the particle is larger than that of the medium, the particles migrate toward the region of a higher electric field strength. For a charged colloidal particle suspended in liquid medium, the electric polarizability of both the particle and its surrounding medium vary with frequency. The relative polarizability of the particle to the medium may change sign at a specific frequency, often referred to as the DEP crossover frequency, at which the direction of DEP force acting on the particle reverses.[3–5] In this case, the crossover frequency reflects the nature of the dielectric relaxation of the particle with its associated electrical double layer at the particle-liquid interface and the dielectric properties of the medium. Understanding the molecular mechanism of DEP crossover is important because it is of interest to particle manipulation in microfluidic environments, such as bio-separation and cell sorting.[6–10]

Both theoretical[11–15] and experimental[3,10,16] studies have been conducted to understand the frequency-dependent dielectrophoretic behavior and the crossover phenomenon of micro- and nano-particles in suspensions. As the intrinsic electrical polarizability of dielectric particles is much smaller than that of the aqueous medium, the relaxation of the electric double layer is believed to have a dominant effect. As proposed by Schwarz, the low-frequency dielectric relaxation mechanism is mainly due to electric double layer diffusion.[4,15–17] Although some success has been achieved to account for the effect of the particle sizes, a significant discrepancy lies between the measured DEP crossover frequency and the relaxation frequency predicted by Schwarz theory.[4,10,18,19]

To investigate the diffusion theory by Schwarz and its applicability in explaining experimental data, we apply the technique of optical tweezers-based DEP force spectroscopy on a single colloidal particle. In contrast to particle image velocimetry, where the macroscopic behavior of a particle ensemble is studied,[18,20] single-particle force detection in conjunction with amplitude modulation and lock-in detection techniques allows us to avoid the complication effects due to electrokinetic mechanisms, such as electrophoresis and AC electroosmosis,[19,21–23] so that only the effects due to DEP are measured. The technique was introduced in

our previous research on DEP-based lab-on-a-chip applications and was used for measurements of the DEP crossover frequency of individual micro colloidal particles[3] as well as that of clusters formed by geometrical stacking of spheres.[4] Those studies did not fully address the underlying mechanisms that would confirm the dielectric response function of charged colloidal particles as suggested by Schwarz.[15] In this single-particle study, we aim to examine the counterion relaxation of individual charged colloidal particles and to provide the microscopic foundation of the bulk dielectric response function as established by Schwarz.[15] Here, we measure the DEP crossover frequency of an individual particle as a function of particles radius, medium viscosity, and temperature. Meanwhile, we solve the Clausius-Mossotti relation with the dielectric response function demonstrated in the Schwarz theory for a charged sphere suspended in a low-salt aqueous solution and compare the trend of theoretical predictions to measured crossover frequencies.

II. THEORETICAL BACKGROUND

The DEP force, F_{DEP} on a spherical particle is proportional to the gradient of the magnitude of electric field squared[2]

$$F_{DEP} = 2\pi r^3 \varepsilon_0 \varepsilon_m Re\{K(\omega)\} \nabla |\bar{E}|^2,$$ (1)

where r is the particle radius, ε_0 is the vacuum permittivity, ε_m is the medium permittivity, and ω is the angular frequency of the external AC electric field $\bar{E}$. The direction of DEP force is determined by the sign of the real part of the Clausius-Mossotti factor, $Re\{K(\omega)\}$.[2] For a spherical particle

$$K(\omega) = \frac{\tilde{\varepsilon}_p(\omega) - \tilde{\varepsilon}_m(\omega)}{\tilde{\varepsilon}_p(\omega) + 2\tilde{\varepsilon}_m(\omega)},$$ (2)

where $\tilde{\varepsilon}_p$ and $\tilde{\varepsilon}_m$ are the complex permittivities of the particle and the suspending medium, respectively. Complex permittivity as a function of frequency is defined as $\tilde{\varepsilon}(\omega) \equiv \varepsilon - j(\sigma/\omega)$, where ε is the real part of the permittivity, $j \equiv (-1)^{1/2}$, and σ is the conductivity; both ε and σ are frequency dependent.

According Eq. (2), a reversal of the direction of the DEP force occurs at the frequency when the real part of the Clausius-Mossotti factor is zero. By solving the relation at the DEP crossover frequency f_{DEP}[9,24]

$$f_{DEP} = \frac{\omega_{DEP}}{2\pi} = \frac{1}{4\pi^2} \sqrt{\frac{(\sigma_p - \sigma_m)(\sigma_p + 2\sigma_m)}{(\varepsilon_m - \varepsilon_p)(\varepsilon_p + 2\varepsilon_m)}}.$$ (3)

By omitting the intrinsic permittivity and conductivity of the colloidal particle ε_{p0} and σ_{p0}, which are orders of magnitude lower than the typical values associated with the electric double layer surrounding the particle in an aqueous medium, ε_{DL} and σ_{DL}, we rewrite Eq. (3) for submicron sized colloids in water

$$f_{DEP} = \frac{1}{4\pi^2} \sqrt{\frac{(\sigma_{DL} - \sigma_w)(\sigma_{DL} + 2\sigma_w)}{(\varepsilon_w - \varepsilon_{DL})(\varepsilon_{DL} + 2\varepsilon_w)}}.$$ (4)

Schwarz derived the double layer's complex permittivity to be

$$\tilde{\varepsilon}_{DL} = \frac{1}{1 + j\omega\tau_\alpha} \frac{e^2 r \delta_0}{k_B T},$$ (5)

where τ_α is the α-relaxation time, ω is the angular frequency, e is the elementary charge, and δ_0 is the surface density of charge (ions per unit area). By rewriting the complex permittivity in the traditional form, we have

$$\tilde{\varepsilon}_{DL} = \varepsilon_{DL} - j\frac{\sigma_{DL}}{\omega} = \frac{1}{1+\omega^2\tau_\alpha{}^2}\frac{e^2r\delta_0}{k_BT} - \frac{j}{\omega}\frac{\omega^2\tau_\alpha}{1+\omega^2\tau_\alpha{}^2}\frac{e^2r\delta_0}{k_BT},$$

(6)

where $\varepsilon_{DL} = \frac{1}{1+\omega^2\tau_\alpha{}^2}\frac{e^2r\delta_0}{k_BT}$ and $\sigma_{DL} = \frac{\omega^2\tau_\alpha}{1+\omega^2\tau_\alpha{}^2}\frac{e^2r\delta_0}{k_BT}$.

By plugging Eq. (6) in Eq. (4), we rewrite the equation in the form of the angular frequency

$$\omega = \frac{1}{2\pi}\sqrt{\frac{\left(\dfrac{\omega^2\tau_\alpha}{1+\omega^2\tau_\alpha{}^2}\dfrac{e^2r\delta_0}{k_BT} - \sigma_w\right)\left(\dfrac{\omega^2\tau_\alpha}{1+\omega^2\tau_\alpha{}^2}\dfrac{e^2r\delta_0}{k_BT} + 2\sigma_w\right)}{\left(\varepsilon_w - \dfrac{1}{1+\omega^2\tau_\alpha{}^2}\dfrac{e^2r\delta_0}{k_BT}\right)\left(\dfrac{1}{1+\omega^2\tau_\alpha{}^2}\dfrac{e^2r\delta_0}{k_BT} + 2\varepsilon_w\right)}}.$$

(7)

Double layer diffusion of colloidal particles in aqueous suspensions is modeled by Schwarz's theory as a process of low-frequency dielectric dispersion (α-relaxation), where the counterion distribution is polarized by the external AC field and forms a concentration gradient along the particle surface. The relaxation time is determined by the diffusion rate of the counterion. In our experiments, particle sizes were larger than the Debye length (λ_D), which was calculated to be $\lambda_D \sim 200\,nm$ in de-ionized water in equilibrium with atmospheric CO_2.[25] For micron and submicron particles that satisfy $r \gg \lambda_D$, namely, the particle size is much larger than the Debye length, α-relaxation time is[10]

$$\tau_\alpha = \frac{1}{\omega_\alpha} = \frac{r^2}{2D},$$

(8)

where r is the particle radius, and D is the counterion diffusion coefficient dominated by proton[26,27] in this study.

To examine the effect of diffusion on the double layer relaxation and its relation to DEP force crossover, we measure the DEP crossover frequency as a function of particle size, medium viscosity, and temperature, and compare experiments with predictions given by Eq. (7). As solved for submicron particles in the Appendix, we express the angular DEP crossover frequency as a function of α-relaxation frequency. Towards a qualitative analysis, we derive the first order approximation as shown in Eq. (A5).

III. MATERIALS AND METHODS

A. Device fabrication

Measurement of DEP motion requires a non-uniform electric field. We achieved the condition by fabricating a pair of gold-film microelectrodes on a glass substrate as shown in Fig. 1. Gold on titanium (Au/Ti, 200 nm/10 nm) microelectrodes were deposited on a microscope cover glass (Fisher Scientific Inc., 12–545-J 22X60–1) using standard photolithography.[4,25] Each electrode was $100\,\mu m$ wide and $1\,cm$ long, and with an angle of $90°$ at pointed tip. The gap between the electrode tips was $28\,\mu m$. For an applied peak-to-peak voltage (Vpp $= 3$ V), the electric field is in the order of 10^5 V/m and the gradient of the field squared in the order of 10^{16} V^2/m^3, at a location along the midline connecting the two tips, $4\,\mu m$ from tip and $0.2\,\mu m$ above the film electrodes.

B. Sample preparation

We prepared a low-concentration (0.001% w/v) colloidal suspension of polystyrene latex spheres in de-ionized water for each of the seven different radii: 370, 445, 500, 650, 750, 1050,
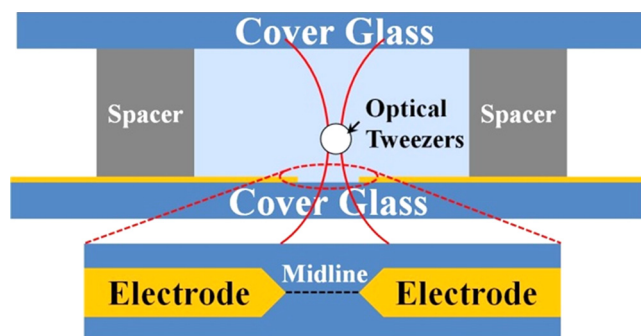
FIG. 1. A schematic side view (top figure) and an enlarged top view (bottom figure) of sample chamber and the gold-film electrodes. The thin layers of yellow strips in the top figure are 200 nm thick films of gold-on-chromium electrodes deposited on the cover glass. A polystyrene bead (white circle in the top figure) is shown, held by optical tweezers at a location a few microns above cover glass and near the midpoint between the two electrodes.

and 1450 nm (Thermo Scientific, 5000 Series). At such low particle concentration, we could isolate and trap an individual particle as force probe without the interference of other particles in the vicinity for each measurement. We used de-ionized (DI) water as solvent and waited for the suspension to equilibrate at atmospheric CO_2 before measurement. The electrical conductivity of the liquid medium, with conductivity due primarily to the ionized carbonic acid, was in the range of 2–3 μS/cm. We did not include salt dependent results in this paper, knowing addition of salts in solution introduces a variety of ionic species. Each species has a different electrophoretic mobility, making our quantitative study of counterion relaxation difficult. As shown in the schematic diagram in Fig. 1, a scotch tape was used as spacer to create a $\sim$100 μm thick chamber that holds approximately 20 μl of sample above the electrodes plane. The sample was confined and vacuum grease-sealed between the two thin glass substrates (12–545-101 22CIR-1, Fisher Scientific).

C. Optical tweezers-based force sensor

Optical tweezers were used to trap individual particles by a focused 1064 nm laser beam using a high numerical aperture microscope objective lens (Olympus PlanFluo, 100X, N.A. = 1.3). We tracked the particle motion with a 980 nm laser beam that was fixed in space and shined through the particle. Movements of the particle can be detected by the projection of the diffracted 980 nm beam on a quadrant photodiode.[28,29] Fig. 2 is a schematic of the optical tweezers-based single-particle force transducer and detector. We used optical tweezers to trap a particle along the midline connecting the electrode tips. The particle was positioned near the center between the electrode tips to minimize the influence from AC electroosmosis, which, the same as DEP, is also E^2 dependent.[25] We use the same design principals described in details before[3,4] to determine the DEP crossover frequencies. Here, a 3 Vpp AC at radial frequencies was applied across the pair of electrodes, creating an electric field gradient. The DEP force pulls the optically tapped particle (white circle) from the center (red dot) of the optical tweezers, as shown in Fig. 2. According to Eq. (1), the DEP force is proportional to the square of the
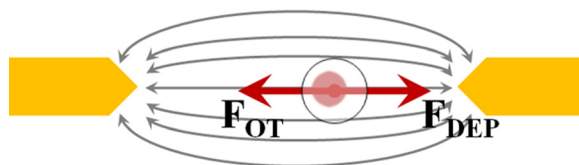

FIG. 2. Top view of force detection by the optical tweezers. The center of a polystyrene bead (white circle) deviates from the focal point of the optical tweezers within the trapping volume (pink disk) under the effects of the DEP force. DEP force is selectively measured by the lock-in amplifier with reference frequency at twice the AM frequency to the applied AC electric field.

AC electric field. By applying amplitude modulation (AM) to the electric field and detecting at twice the modulation frequency using a lock-in amplifier (Stanford Research SR830), we extracted the field-squared-dependent force. Since the particle was located far from the electrodes, the effect due to AC electroosmosis was negligible, and the measured force was dominated by the DEP force.[25] The AM frequency was two orders of magnitude lower than the DEP frequency to avoid crosstalk with the DEP frequency and to allow the particle to be responsive to the modulated DEP force such that the particle motion is detectable. We identify the DEP crossover frequency to be the frequency at which the magnitude of the particle motion approaches zero and the phase of the particle motion switches by 180°.[3,4]

IV. EXPERIMENTAL RESULTS AND DISCUSSION

At a fixed sample temperature at 27 °C, the DEP crossover frequencies as a function of the particle size are shown in Fig. 3. The least-squared fitting, indicated by the red solid line, gives an exponent of -1.7 ± 0.1, which is in agreement with a recently reported -2 power dependence for micron-sized PMMA particles.[4] We evaluated the DEP crossover frequency using Eq. (A5) and plotted those together with the experimental data. The comparison between the experiments and the theoretical calculations on the size-dependence is shown in Fig. 3. Our measurements showed that the DEP crossover frequency of micro particles does not change with medium conductivity in the low conductivity regime (2–780 μS/cm). These results are in agreement with the experiments and the theoretical models presented by Ermolina and Morgan[17] and by Basuray et al.[30]

To further examine how the counterion diffusion within the Debye layer affects the DEP crossover, we studied the DEP crossover frequency of a 750 nm particle as a function of medium viscosity by suspending the particle in different concentrations of glycerol solutions. By adding 0% to 40% of glycerol by volume to the de-ionized water at 27 °C, we varied the medium viscosity from 0.8 mPa s to 8 mPa s.[31,32] The medium permittivity is in the range of 60 and 78, and conductivity is in the range ~2 μS/cm. The experimental results of the DEP crossover frequency as a function of the reciprocal of the medium viscosity are shown as black squares in Fig. 4. The inverse proportionality confirms the diffusive nature of dielectrophoretic relaxation. To compare with the results reported by Ermolina and Morgan[17] their data on particles in the radius range of 55–230 nm showed the DEP crossover frequency stopped varying with the viscosity increase beyond 2 mPa s, which was a phenomenon not observed in our experiments with micro particles. Our result implies the micro-sized particle's DEP crossover
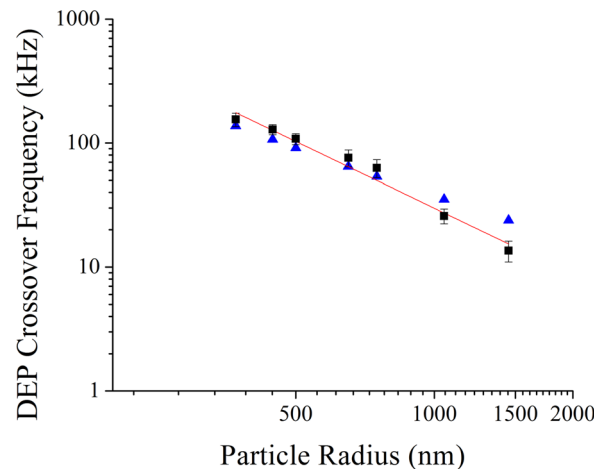

FIG. 3. DEP crossover frequency as a function of particle size in de-ionized water at 27 °C. The measurements are shown by the black squares with error bars and theoretical calculations by the blue triangles. Mean value and standard deviation were obtained by repeating 3 times of measurements. The logarithmic fitting of the data, shown by the red solid line, gives the best fit with an exponent of -1.7 ± 0.1.
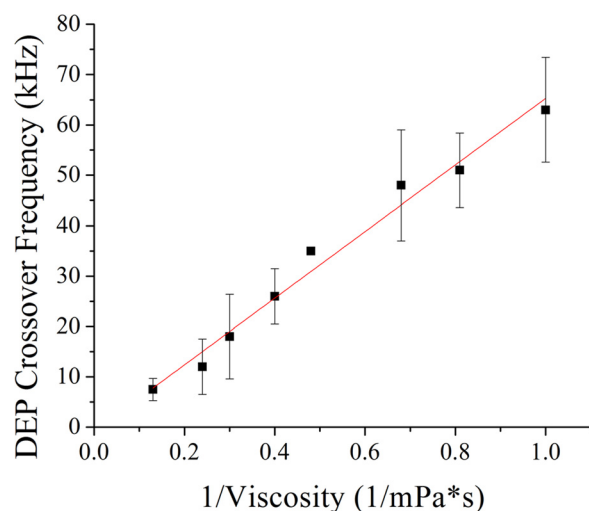
FIG. 4. DEP crossover frequency of a 750 nm polystyrene particle as a function of 1/viscosity at 27 °C. The measurements are shown by the black squares with error bars. Mean value and standard deviation were obtained by repeating 3 times of measurements. The linear fitting of the data is shown by the red solid trend-line.

frequency is inversely proportional to the α-relaxation time and indicates that DEP crossover phenomenon in low viscosity medium is contributed primarily by the ionic diffusion in the counterion double layer.

We measured the temperature dependence of the DEP crossover frequency of a 750 nm particle in de-ionized water. The corresponding medium viscosity was recorded to account for its variation as a function of temperature. In Fig. 5, the crossover frequency multiplied by medium viscosity is an increasing function of the temperature. The major difference between the measured DEP crossover frequencies and the theoretical predictions in the higher temperature range can potentially be explained as a consequence of activated counterion migration along the particle surface as proposed by Schwarz,[15] that the surface charge density may depend on the temperature. However, the limited temperature range of our experiment was too narrow to give an accurate determination of the activation process.
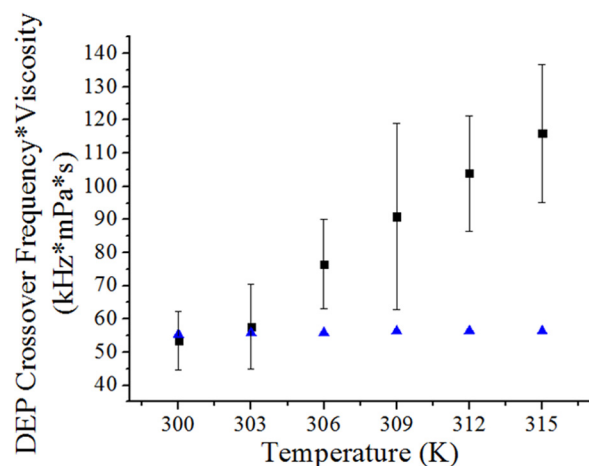

FIG. 5. DEP crossover frequency multiplied by viscosity of a 750 nm polystyrene particle as a function of the temperature. The results in de-ionized water of temperature variance between 27 °C and 42 °C are labeled on black squares with error bars and theoretical calculations by the blue triangles. Mean value and standard deviation were obtained by repeating 3 times of measurements.

V. CONCLUSIONS

By examining the dependences of particle size, medium viscosity, and temperature, we conducted a comprehensive study on low-frequency dielectric response of charged colloidal particles in aqueous suspensions and compared the results to Schwarz's theory. We focused on experimental data for DI water, where protons are the dominant ionic species that is responsible for the double layer polarizability. Comparing our experimental data and the theoretical model by Schwarz for low-frequency dielectric response for micron-sized particles in low conductivity medium, we conclude that the crossover phenomenon in DEP is contributed primarily by the ionic diffusion in the electric double layer. While our viscosity dependent data suggested the diffusive nature of counterion double layer, the temperature data hinted the possibly of additional effects. Those may include temperature-enhanced hopping of protons between the host water molecules[32] and thermal activation of surface-bound ions beneath the diffuse double layer.[14] In order to derive more definitive explanations, conducting experiments in a wider temperature range, altering surface-bound ions, and modifying the medium conductivity will be necessary. Accurate measurements of a single particle DEP crossover frequency, as promised by employing optical tweezers-based force spectroscopy, enabled a close examination of the role of double layer diffusion in DEP.

ACKNOWLEDGMENTS

APPENDIX: DEVIATION OF THEORETICAL DEP CROSSOVER FREQUENCY

We start from Eq. (7) and use the values $\varepsilon_w = 78\varepsilon_0$ and $\sigma_w \cong 2$ μS/cm to calculate the crossover angular frequency. We divide both the numerator and denominator in the square root by ε_0 and define $x = \frac{e^2 \delta_0 r}{\varepsilon_0 k_B T}$

$$\omega = \frac{1}{2\pi} \sqrt{\frac{\left(\frac{\omega^2 \tau_\alpha}{1+\omega^2 \tau_\alpha^2} x - \frac{\sigma_w}{\varepsilon 0}\right)\left(\frac{\omega^2 \tau_\alpha}{1+\omega^2 \tau_\alpha^2} x + 2\frac{\sigma_w}{\varepsilon 0}\right)}{\left(78 - \frac{1}{1+\omega^2 \tau_\alpha^2} x\right)\left(\frac{1}{1+\omega^2 \tau_\alpha^2} x + 156\right)}}. \tag{A1}$$

We use a 750 nm particle at 25 °C as an example. According to Eq. (8), $\tau_\alpha = \frac{r^2}{2D} = \frac{(750 \times 10^{-9})^2}{2 \times 9.31 \times 10^{-9}}$ $= 3 \times 10^{-5}$, where $D = 9.31 \times 10^{-9}$ m^2/s is the free proton diffusivity at room temperature.[26,27]

For simplicity of estimating the value of $\omega\tau$, we assume a typical DEP crossover frequency to be on the order of 50 kHz. $\omega^2 \tau_\alpha^2 \cong (3 \times 10^{-5})^2 (2 \times 50 \times 10^3 \pi)^2 = 89$. Therefore

$$\frac{\omega^2 \tau_\alpha}{1+\omega^2 \tau_\alpha^2} \cong \frac{\omega^2 \tau_\alpha}{\omega^2 \tau_\alpha^2} = \frac{1}{\tau_\alpha}. \tag{A2}$$

We then use a typical surface charge density for a polymer colloid particle $\delta_0 = 10^{16}$/m^2 and $\sigma_w \cong 2$ μS/cm

$$\frac{\omega^2 \tau_\alpha}{1+\omega^2 \tau_\alpha^2} x \cong \frac{x}{\tau_\alpha} = (3 \times 10^{-5})^{-1} \times 5238 = 1.7 \times 10^8 \quad \text{and} \quad \frac{\sigma_w}{\varepsilon_0} = \frac{2 \times 10^{-4}}{8.854 \times 10^{-12}} = 2.2 \times 10^7$$

$$\frac{\omega^2 \tau_\alpha}{1+\omega^2 \tau_\alpha^2} x > \frac{\sigma_w}{\varepsilon_0}. \tag{A3}$$

Given the approximations in Eqs. (A2) and (A3), we reduce the equation to

$$\omega = \frac{1}{2\pi} \sqrt{\frac{\left(\frac{x}{\tau_\alpha}\right)\left(\frac{x}{\tau_\alpha}\right)}{\left(\frac{\varepsilon_w}{\varepsilon_0} - \frac{x}{\omega^2 \tau_\alpha{}^2}\right)\left(\frac{x}{\omega^2 \tau_\alpha{}^2} + 2\frac{\varepsilon_w}{\varepsilon_0}\right)}}. \tag{A4}$$

For submicron particles, we derive the angular DEP crossover frequency $\omega = \omega_{\mathrm{DEP}}$ as

$$\omega_{\mathrm{DEP}} = \frac{1}{4\pi\tau\alpha} \sqrt{\frac{4\frac{\varepsilon_w}{\varepsilon_0}\pi^2 x + x^2 + \sqrt{144\left(\frac{\varepsilon_w}{\varepsilon_0}\right)^2 \pi^4 x^2 + x^4 + 8\frac{\varepsilon_w}{\varepsilon_0}\pi^2 x^3}}{\frac{\varepsilon_w}{\varepsilon_0}}}. \tag{A5}$$

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